AD696136

METHODS FOR OBTAINING LASER ACTION

AT WAVELENGTHS SHORTER THAN 3000 Å

SEMI-ANNUAL STATUS REPORT NO. 1

for the period

1 March 1969 - 31 August 1969

Office of Naval Research
Contract N00014-67-A-0112-0033
NR 016-107
Principal Investigator: R. H. Pantell

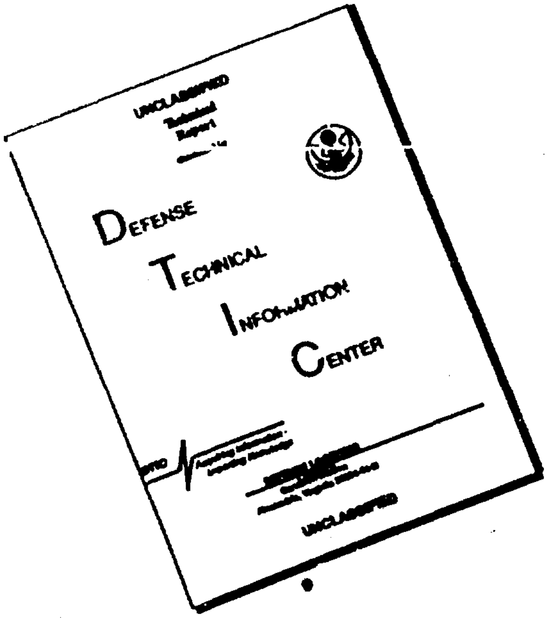
M. L. Report No. 1790

September 1969

Microwave Laboratory
W. W. Hansen Laboratories of Physics
Stanford University
Stanford, California

12

DISCLAIMER NOTICE



THIS DOCUMENT IS BEST QUALITY AVAILABLE. THE COPY FURNISHED TO DTIC CONTAINED A SIGNIFICANT NUMBER OF PAGES WHICH DO NOT REPRODUCE LEGIBLY.

METHODS FOR OBTAINING LASER ACTION
AT WAVELENGTHS SHORTER THAN 3000 Å

OBJECTIVE

The general objective of this research program is to investigate methods for achieving laser action at wavelengths less than 3000 Å.

INTRODUCTION

There are many reasons why it would be desirable to develop lasers in the ultraviolet and X-ray regions of the specturm. Possible applications include fluorescence studies and devices, short wavelength holography, band structure studies in solids, atomic and molecular spectroscopy, biological research, medical applications, X-ray transmission, and high energy physics research.

Unfortunately, as frequency increases it becomes pregressively more difficult to achieve laser action by the standard process of population inversion. The reason for the difficulty is that with a given pump power density the gain at the laser wavelength decreases rapidly with increasing frequency. Thus, it is desirable to investigate other methods for achieving coherent stimulated emission at short wavelengths.

We are investigating two techniques, which are applicable to short wavelength laser action. The first is two-step laser action. The

second is a tunable Cerenkov laser. These research projects are described below.

PROJECT I: <u>TWO-STEP LASER ACTION</u> (D. L. Hecht, R. H. Pantell)

A. DESCRIPTION

The principal barrier to ordinary laser action in the ultraviolet is that the pump power required to overcome spontaneous emission is very large. The minimum required pumping power goes up in proportion to at least the third power of the laser frequency.

We are investigating a two-step pumping technique which may overcome this difficulty. A typical desired energy level configuration is illustrated in Fig. 1.

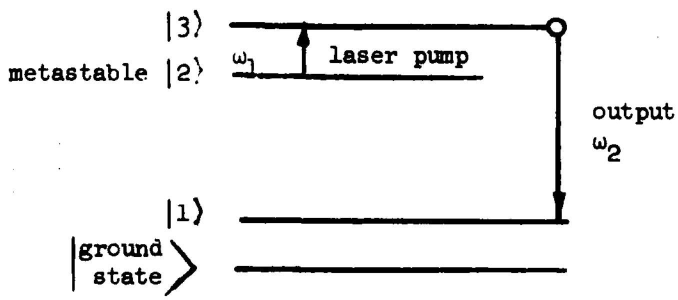


FIG. 1--Double pumped laser action.

The key idea is to first build up a considerable population in the long lived metastable energy level $|2\rangle$. This may be accomplished by conventional pumping which need not reach a very high peak power and

may profitably last over the lifetime of state $|2\rangle$. In this way state $|2\rangle$ acts as an energy store. Then a short intense laser light pulse which is resonant on the $|2\rangle - |3\rangle$ transition is applied to the active medium. With this intense pulse at ω_1 stimulated emission may be achieved at ω_2 .

There are two possible mechanisms for inducing a coherent output on the $|3\rangle - |1\rangle$ transition frequency ω_2 . The first of these is the stimulated resonant anti-Stokes process involving levels $|2\rangle$, $|3\rangle$ and $|1\rangle$. Secondly, the laser pulse may populate level $|3\rangle$ to obtain significant population inversion with respect to level $|1\rangle$. This would be a laser-pumped-laser in which the output frequency is much higher than the input frequency.

B. PRESENT STATUS

1. Theoretical

We have developed equations for the exponential gain at the output frequency ω_2 for the two processes in a system with the energy configuration of Fig. 1.

The exponential gain Γ_{AS} (meters^{-1}) for resonant anti-Stokes stimulated emission is given by Eq. 1

$$\Gamma_{AS} = \left[\frac{\pi^4 e^4}{4 c^2 m^2 \epsilon_0^2 h}\right] \frac{N_2}{\eta_{23}\eta_{13}} \frac{f_{23} f_{13} g_{12} g_{23} g_{13}}{\omega_{23}} S_{23} \text{ (MKS)} \qquad (1)$$

where

e = electron charge

m = electron rest mass

c = speed of light in vacuo

ϵ_0 = free space permittivity

h = Planck's constant

N_i = population of level i (meters^{-3})

ω_{ij} = radian frequency of light for energy difference between states i and j

η_{ij} = index of refraction of active medium at ω_{ij}

f_{ij} = oscillator strength between states i and j

g_{ij} = value of normalized line shape factor on resonance $= g_{ij}(\omega_{ij})$

S_{23} = power density of pump laser (WATTS/meter^2)

Evaluating the universal constants gives

$$\Gamma_{AS} = 4.143 \times 10^{24} \times \frac{N_2}{\eta_{23}\eta_{13}} f_{23} f_{13} \frac{g_{21} g_{23} g_{13}}{\omega_{23}} S_{23} \text{ (MKS)} \quad . \qquad (2)$$

The corresponding result for two-step pumping of ordinary laser action is:

$$\Gamma_{LASER} = \left(\frac{\pi e^2}{8hc^2\epsilon_0 m}\right) N_2 g_{31} g_{23} f_{23} S_{23} \lambda_{31}^2 \lambda_{21} \quad (MKS) \qquad (3)$$

where λ_{ij} is the wavelength corresponding to ω_{ij} .

Evaluating the universal constants gives

$$\Gamma_{LASER} = 2.10 \times 10^{19} N_2 g_{31} g_{23} f_{23} S_{23} \lambda_{31}^2 \lambda_{21} \quad (MKS) \quad . \qquad (4)$$

A feasibility criterion for these processes is that Γ should exceed 1 meter^{-1} .

2. Experimental

As a prototype system of this form we have been investigating fluorene, an aromatic hydrocarbon, $C_6H_4CH_2C_6H_4$. Its energy level diagram is shown in Fig. 2, which may be compared with Fig. 1.

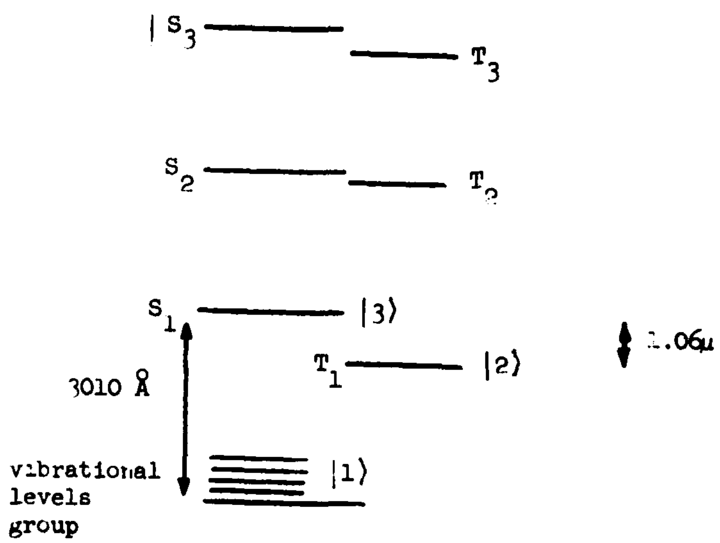


FIG.--Fluorene energy level configuration.

The lowest excited singlet state S_1 corresponds to level $|3\rangle$ of the general system. The unpopulated vibrational levels above the ground in fluorene correspond to level $|1\rangle$. The lowest triplet state T_1 corresponds to level $|2\rangle$.

Fluorene was chosen as a prototype medium because of the following favorable properties. The oscillator strength for the $|3\rangle - |1\rangle$ fluorescence transition is high, $f_{31} = 0.13$. The triplet state lifetime in a rigid host medium is very long — about 5 seconds. The energy level spacing between S_1 and T_1 is coincident with the Nd^{+3} glass laser output, providing an excellent second step pumping source. The output wavelength for fluorene is 3000 Å . However, there are some significant negative factors. The linewidth of the $|3\rangle - |1\rangle$ transition is rather broad (more than 1000 cm^{-1}). Also, a critical question is that the parameters of the $|2\rangle - |3\rangle$ coupling are unknown.

The experimental program has consisted of the following:

1. The highest purity commercial fluorene was obtained and repurified to remove dibenzfuran and other impurities.

2. Fluorene fluorescence and phosphorescence spectra were obtained at 77°K in EPA (alcohol-ether) solution.

3. Special triple jacket quartz cells were constructed to contain fluorene in rigid EPA glass solution at liquid nitrogen temperature. Solutions made with dry EPA were successfully frozen into transparent glass. However, the glass was extremely fragile and unsuitable for laser applications.

4. We obtained special high purity PMMA (polymethylmethacrylate) plastic doped with our purified fluorene. The concentrations

ranged from 10^{-3} to 10^{-2} mole/liter. The plastic had good optical quality. We measured the absorption spectra of the intrinsic polymer and doped polymer from 2600 Å to 16,000 Å. The intrinsic polymer transmitted down to about 2775 Å. For comparison commercial plexiglass cuts off at 3500 Å. The plastic is extremely convenient to work with compared to crystals and glasses and appears to be a good laser host material for the visible and near ultraviolet. The fluorene spectrum appears to be the same in PMMA as in EPA. The phosphorescence lifetime is about 5 seconds.

5. A Q-switched Nd^{+3} glass laser was built and operated with a 6" × 3/8" Nd^{+3} glass rod. The output power is several megawatts. A cavity was built with 4 linear flashlamps to achieve the first step of pumping. The maximum input power is 2000 joules in 300 microseconds which should yield about 100 joules of ultraviolet light for the first step pumping. The two flash cavities are coupled by a dual triggering system with variable delay. See Figure 3.

6. We unsuccessfully attempted to observe ultraviolet output stimulated by the infrared laser incident on a 10 cm × 1/2" doped plastic rod in the flash cavity. At high flash lamp pumping power the plastic developed internal fractures. It would be better to pump with a series of smaller flash lamp pulses. Also, if we focused the secondary pumping laser with a 50 cm lens, the plastic was damaged. This limits the pumping intensity S_{23}.

Our inability thus far to generate stimulated ultraviolet from the metastable level in fluorene indicates that the $|2\rangle - |3\rangle$ transition is extremely weakly coupled. This indicates that this transition may be like the $|2\rangle - |1\rangle$ transition which is so weak that a 5 second

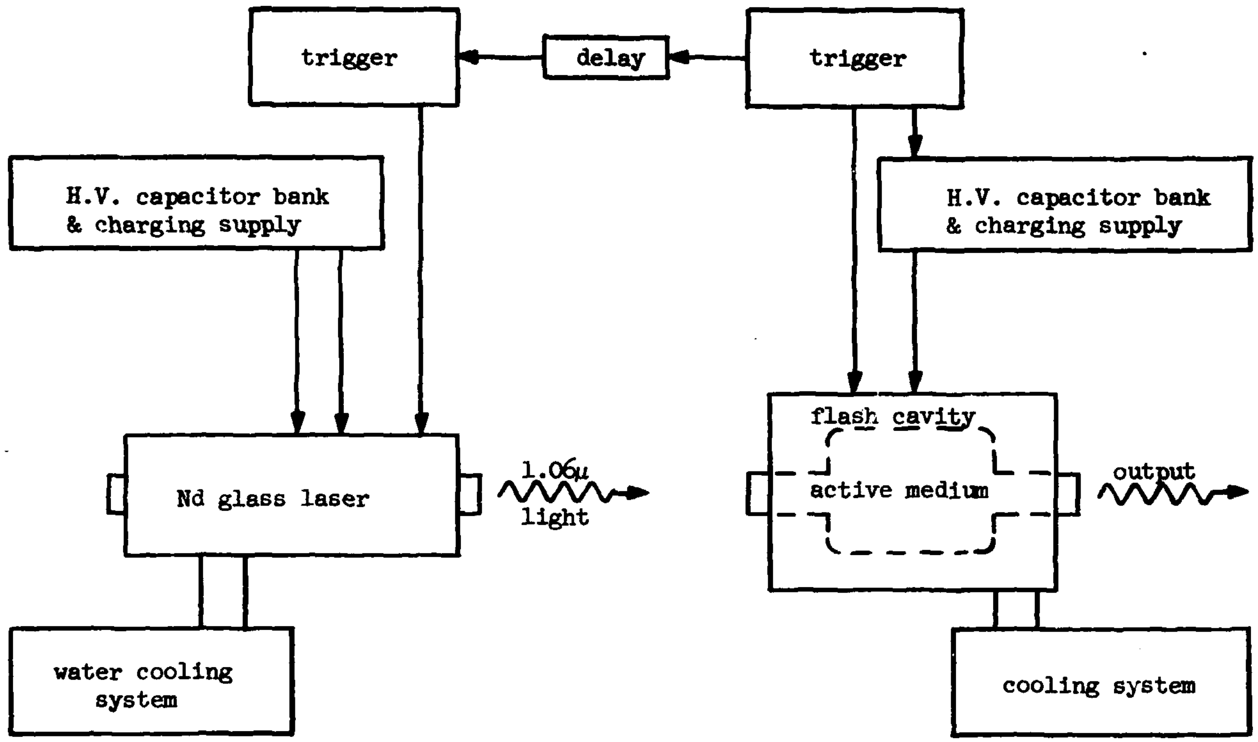


FIG. 3--Double pumped fluorene laser system.

metastable lifetime for state $|2\rangle$ results. If the $|2\rangle - |3\rangle$ transition is as weak as the $|2\rangle - |1\rangle$ transition, f_{23} could be as low as 10^{-10}. If this is the case, it would be far too low to enable stimulated emission at feasible pumping powers.

C. FUTURE WORK

Our plans for future work include the following:

1. Determining if anything can be done to enhance the $|2\rangle - |3\rangle$ coupling in fluorene.

2. Investigating fluorene as a single step pumped dye laser. No dye laser has been operated as far into the ultraviolet as 3000 Å. A fluorene dye laser should be tunable from 3000 Å to 3200 Å by adjusting the optical cavity.

3. Investigation of other materials as the active medium.

4. Further theoretical development and comparison of the two stimulated emission processes.

PROJECT II: TUNABLE CERENKOV LASER (M. Piestrup, H. Puthoff)

A. DESCRIPTION

This project is directed toward the design and construction of a new type of laser device, an electron pumped Cerenkov laser. Preliminary theoretical and experimental work indicates that exploitation of the Cerenkov effect as a source of coherent laser radiation may be possible.

When high energy electrons pass through a material at a velocity exceeding the speed of light in the material, a broad spectrum of electro-

magnetic radiation is generated by the Cerenkov effect. A laser employing the Cerenkov effect would have the outstanding characteristics of (1) being broadly tunable from the far infrared to the far ultraviolet and (2) possibly providing a technique for extending laser technology into the X-ray region of the spectrum.

B. PRESENT STATUS

1. Theoretical

The spontaneous parametric fluorescence power per unit bandwidth has been calculated and compared with that produced by optical parametric devices and found to exceed the latter by a factor of more than 10^9 under typical operating conditions. This indicates that accelerator generated Cerenkov light under appropriate conditions may prove to be a useful source of light for, e. g., spectroscopic applications.

Gain and threshold calculations have been carried out to determine whether stimulated Cerenkov radiation in a laser mode is possible. At this point the tentative results support the feasibility of this mode of operation.

2. Experimental

Since June several experiments have been carried out at the Stanford High Energy Physics Laboratory accelerator in which the accelerator electron beam irradiated a special radiation-resistant quartz sample which was coated with dielectric mirrors to form an optical cavity. Brilliant tunable light has been observed which spanned the entire visible spectrum.

C. FUTURE WORK

1. Further theoretical work is planned to verify the present results.

2. Experiments are planned to determine the behavior of the observed radiation, especially to determine whether the emission is stimulated and coherent. Success would indicate a major breakthrough in laser technology.

ACKNOWLEDGEMENTS

We would like to extend our sincere appreciation and thanks to the following:

Dr. N. Bortnick and Dr. E. Reed Lang of the Rohm and Hass Co. Research Laboratory for providing us with excellent plastic samples, Dr. R. Haugland[1] of Stanford for purification of our fluorene, Dr. J. Yguerabide[2] of Stanford for fluorescence and phosphorescence measurements on fluorene and numerous helpful discussions, J. M. Yarborough of Stanford for contributions to the experimental program, and Randy Whitney and George Rothbart of the High Energy Physics Laboratory for their cooperation in the use of the 500 MeV accelerator.

PROJECT STAFF

Principal Investigator: Prof. R. H. Pantell

Research Associate: Dr. H. Puthoff

Research Assistants: D. L. Hecht and M. Piestrup

---

1 Presently with Syntex

2 Presently with Yale University.